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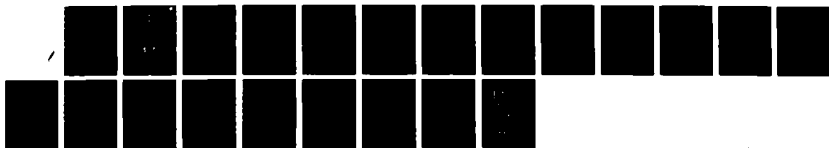
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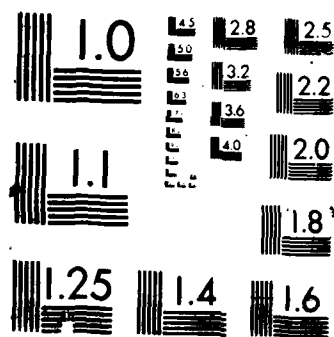
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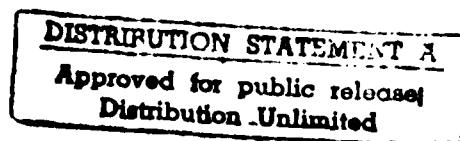
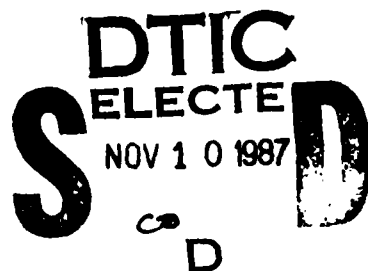
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# THERMOLUMINESCENCE RESPONSE OF $\text{CaF}_2\text{:Mn}$ , $\text{CaF}_2\text{:Dy}$ and $\text{CaSO}_4\text{:Tm}$ TO PROTONS AND ALPHA-PARTICLES

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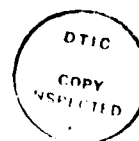


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# THERMOLUMINESCENCE RESPONSE OF $\text{CaF}_2\text{:Mn}$ , $\text{CaF}_2\text{:Dy}$ and $\text{CaSO}_4\text{:Tm}$ TO PROTONS AND ALPHA-PARTICLES

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S. McGowan and R.A. Gravelle  
*Nuclear Effects Section*  
*Protective Sciences Division*



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### ABSTRACT

Proton/gamma and alpha/gamma response ratios have been measured for very fine powders of the thermoluminescent phosphors  $\text{CaF}_2\text{:Mn}$ ,  $\text{CaF}_2\text{:Dy}$  and  $\text{CaSO}_4\text{:Tm}$ . This has been done as part of an evaluation of these phosphors for use in a combined neutron/gamma thermoluminescent dosimeter which is made neutron sensitive by embedding the phosphor powder in an hydrogenous medium. The  $\text{CaF}_2\text{:Mn}$  phosphor has been found to be marginally better than the other two for this purpose. Reduction in the measured proton/gamma response ratio with decreasing proton energy has been attributed primarily to relatively low response near the surfaces of the phosphor particles.

### RÉSUMÉ

Nous avons mesuré les rapports du rendement proton/gamma et alpha/gamma des dosimètres thermoluminescents en poudre très fine ( $\text{CaF}_2\text{:Mn}$ ,  $\text{CaF}_2\text{:Dy}$ ,  $\text{CaSO}_4\text{:Tm}$ ). Nous avons fait l'évaluation de ces luminescents afin de les utiliser dans un détecteur thermoluminescent neutron/gamma combiné. Ce détecteur devient sensible aux neutrons par l'incorporation de la poudre luminescente dans un medium hydrogèneux. Nous avons découvert que le luminescent  $\text{CaF}_2\text{:Mn}$  était supérieur aux deux autres. Nous attribuons la baisse du rapport du rendement mesuré proton/gamma avec diminution de l'énergie des protons à un rendement faible près de la surface des particules luminescentes.

## 1. Introduction

Translating thermoluminescence (TL) response into units of absorbed dose is a problem associated with the use of the thermoluminescent dosimeter (TLD) in diverse radiation fields, such as mixed neutron-gamma fields. TL responses of the detector may depend not only on the photon and neutron energy absorption of the detector's material, but also on the linear energy transfer (LET) of the ionizing particles (Ref. (1) and (2)). Horowitz (3) has reported the effect of batch composition of TL material on the LET dependence of response.

This paper focuses on the dependence on LET of the relative response of three TLD materials:  $\text{CaF}_2\text{:Mn}$ ,  $\text{CaF}_2\text{:Dy}$  and  $\text{CaSO}_4\text{:Tm}$ . These three materials are commonly used in TLD's, because of their high sensitivities and suitable readout temperatures.  $\text{CaSO}_4\text{:Tm}$  powder embedded in polyethylene was investigated at DREO (Ref. (4)) as a combined neutron/gamma TLD, but was found to respond poorly at low neutron energies. Since the neutron response of this TLD is derived from knock-on protons, these proton-response measurements relate directly to the problems with this TLD and the results may lead to a solution.

## 2. Experimental Procedure

The materials used for the measurement of the thermoluminescence (TL) response are:  $\text{CaF}_2\text{:Mn}$ ,  $\text{CaF}_2\text{:Dy}$ , and  $\text{CaSO}_4\text{:Tm}$ . The  $\text{CaF}_2\text{:Mn}$  and  $\text{CaF}_2\text{:Dy}$  were obtained from Harshaw Chemicals and the  $\text{CaSO}_4\text{:Tm}$  was produced in this laboratory (5).

The  $\text{CaF}_2\text{:Mn}$  was investigated in both polycrystalline and powder form while only powders of the other phosphors were tested. The polycrystalline TLD's, which were 3 mm x 3 mm x 0.8 mm in size, were reused many times with no apparent deterioration. Dosimeters were made from the powdered phosphors by retaining the phosphor grains in a matrix of Silastic. A mixture of 5 to 10% powder with the liquid form of the Silastic was deaerated and allowed to set between uniformly-spaced glass plates. These were rotated during setting to prevent settling of the phosphor under gravity. The resulting phosphor/Silastic sheets, which were of thickness between 0.1 and 0.2 mm, were cut into 3 mm x 3 mm samples.

Both of the calcium fluoride powders originally contained grains which ranged up to several hundred micrometers in diameter. After grinding, the larger particles were removed by sedimentation in water. Very fine powders were obtained after sedimentation times of several hours. The  $\text{CaF}_2$ /Silastic samples investigated contained only grains which were less than about  $2\text{ }\mu\text{m}$  in diameter. Phosphor powders of this size are referred to in this report as being "ultrafine". Two grades of  $\text{CaSO}_4:\text{Tm}$  powder were tested using  $\text{CaSO}_4:\text{Tm}$ /Silastic dosimeters made using a "fine" powder, which contained phosphor grains up to  $10\text{ }\mu\text{m}$  in diameter, or using an ultrafine powder. Investigation of the ultrafine powders is relevant to the combined neutron/gamma dosimeter as this grade of phosphor is required for good response to neutrons below a few MeV.

The irradiations were carried out with a  $^{60}\text{Co}$  source, with monoenergetic protons of energies ranging from 0.37 to 2.0 MeV, produced by the DREO Van de Graaff accelerator, and with a 4.3-MeV  $^{241}\text{Am}$  alpha source. Because of the short ranges of the protons and alphas, it was necessary to carry out these irradiations under vacuum. A silicon particle detector was included to measure the proton or alpha fluence.

For the proton irradiations a vacuum chamber was coupled to the end of the accelerator drift tube. The exposure of the TL materials and the proton-beam monitoring were carried out using the "proton chopper" arrangement shown in Fig 1. The TL samples were rotated past a 2-mm slit which defined the proton beam. The proton beam was sampled by the stationary silicon detector when the pin hole passed behind the slit. The  $100\text{-}\mu\text{m}$ -diameter pin hole served to reduce the count rate of proton pulses to a level which could be handled by a pulse-height analyser. Nickel foils were used to reduce the proton energy below 0.8 MeV which was the lowest energy which could be obtained with this accelerator. These were placed over the 2-mm slit. The 0.0001-inch nickel foils were found to reduce proton energies of 1.0 and 0.8 MeV to 0.64 and 0.37 MeV, respectively.

The strength of the  $^{241}\text{Am}$  alpha source was also measured using a silicon detector. The 0.0001-inch nickel foil reduced the 4.3-MeV alphas to 1.4 MeV. A pulse-height analyser was used to record the proton and alpha counts and to determine the energy of the particles after attenuation by the nickel foil.

The TL samples were read with a Harshaw 2000 TL detection system which uses an EMI 9514S photomultiplier tube.

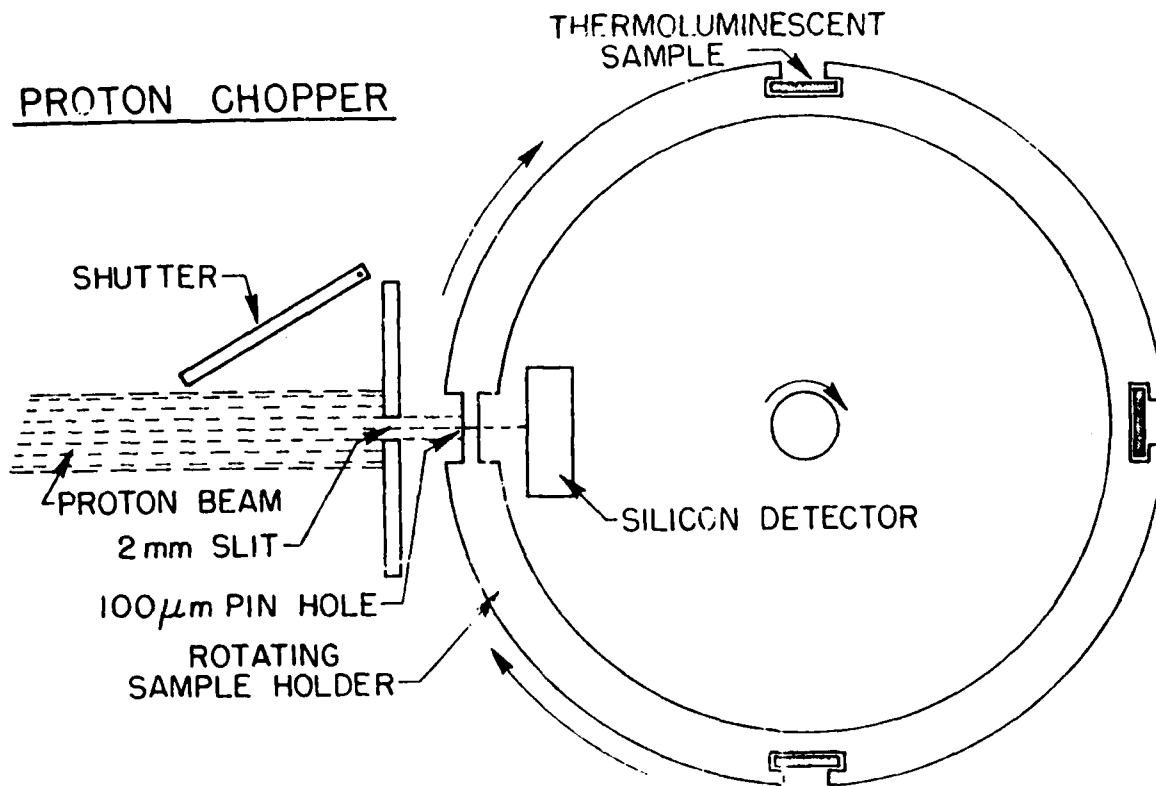


Figure 1: Diagram of the apparatus used for proton irradiation of the phosphor samples. The samples were irradiated as they rotated past the slit defining the proton beam. The proton fluence was measured by the silicon detector which was exposed to the proton beam while the pin hole rotated past the slit.



### 3. Measured TL Response

It is not usually practicable to measure TL response in absolute terms of light output per unit absorbed energy since this requires knowing the absolute sensitivity of the light detection system. It is more common, and usually just as useful, to measure relative sensitivities of various phosphors or of a particular phosphor to different radiation sources. It should be kept in mind that these relative sensitivities can depend on the detection system, since different light detectors have different spectral responses and different phosphors have different spectral emissions. The TL readings are made in some arbitrary units, such as nC, and sensitivities can be expressed in nC/J or nC/(kg cGy). Of course, these units cancel out when sensitivities are measured relative to the sensitivity of some "standard" phosphor or some "standard" radiation source.

#### 3.1 Relative Response of Phosphors to Gammas

The approximate sensitivities of the phosphors investigated are listed in Table I, where the responses (TL emission per energy absorbed) to  $^{60}\text{Co}$  gammas are given relative to that for polycrystalline  $\text{CaF}_2:\text{Mn}$  TLD's. Corrections have been made for optical absorption in the samples. The approximate temperatures  $T_m$  for maximum TL emission are also included.

These sensitivities depend to some extent on the annealing and reading procedures as well as on the method of preparation of the samples. The polycrystalline  $\text{CaF}_2:\text{Mn}$  samples were pre-annealed at  $400^\circ\text{C}$  for 30 min., while the powders in Silastic were pre-annealed at  $275^\circ\text{C}$  for 30 to 60 min. All samples were post-annealed at  $100^\circ\text{C}$  for 30 min. before reading. A blue/green filter (Corning CS4-67) was used in the reader for the  $\text{CaF}_2:\text{Mn}$  and  $\text{CaF}_2:\text{Dy}$  samples and a blue filter (Corning CS5-61) for the  $\text{CaSO}_4:\text{Tm}$  samples to match the wave lengths of the TL emissions.

The two types of  $\text{CaF}_2:\text{Mn}$  polycrystalline TLD's refer to the two groups found to have different optical transmission, type A having the higher optical density. As mentioned earlier, only ultrafine powders of  $\text{CaF}_2:\text{Mn}$  and  $\text{CaF}_2:\text{Dy}$  were investigated with the protons. These had responses to  $^{60}\text{Co}$  gammas which were about 15 and 25% of the respective responses for the unseparated powders of  $\text{CaF}_2:\text{Mn}$  and  $\text{CaF}_2:\text{Dy}$ . Thus, all of these phosphors are seen to have comparable sensitivities which are considerably reduced when the phosphors are converted to very fine powders.

TABLE I

Relative Response to  $^{60}\text{Co}$  Gammas

| <u>Phosphor</u>       | <u>Form</u>                  | <u>T<sub>m</sub>(°C)</u> | <u>Relative Responses</u> |
|-----------------------|------------------------------|--------------------------|---------------------------|
| CaF <sub>2</sub> :Mn  | polycrystalline (Type A)     | 260                      | 1.0                       |
| CaF <sub>2</sub> :Mn  | polycrystalline (Type B)     | 260                      | 0.8                       |
| CaF <sub>2</sub> :Mn  | ultrafine powder in Silastic | 260                      | 0.3                       |
| CaF <sub>2</sub> :Dy  | ultrafine powder in Silastic | 160                      | 0.4                       |
| CaSO <sub>4</sub> :Tm | fine powder in Silastic      | 200                      | 0.8                       |
| CaSO <sub>4</sub> Tm  | ultrafine powder in Silastic | 200                      | 0.4                       |

3.2 Response of CaF<sub>2</sub>:Mn to Proton and Alpha Particles Relative to  $^{60}\text{Co}$

The response  $\eta$  of a TL material to protons (hydrogen ions) or alphas (helium ions) relative to that to  $^{60}\text{Co}$  gamma radiation is a measure of the relative efficiencies of these ions compared with electrons for conversion of absorbed energy into luminescence. This relative response is defined as

$$\eta = \frac{(R/W)_I}{(R/W)_\gamma} \quad (1)$$

where R represents the TL reading and W the absorbed energy. I and  $\gamma$  refer to the ion and gamma radiations.  $\eta$  is the relative efficiency for conversion of the energy of a high-LET particle (ion) into TL compared with that efficiency for a low-LET particle (electron). The TL samples are thick compared with the ion ranges, so that W for the ions is the ion energy times the number of ions absorbed. For  $^{60}\text{Co}$  irradiation W is the product of the dose to the sample and the sample mass. When the sample used was a powder embedded in Silastic it was necessary to take into account the fraction of energy absorbed in the phosphor itself.

Relative response measurements as defined in Eq. (1) were made for protons ( $\eta_p$ ) and alphas ( $\eta_\alpha$ ) for each of the three types of  $\text{CaF}_2\text{:Mn}$  dosimeters included in Sec 3.1. For each type, several proton irradiations were made with one to six samples in each. These were accompanied by gamma irradiations of approximately six samples in order to obtain comparative measurements under the same annealing and reading conditions. The results listed in Table II are averages of ten to twenty proton-irradiated samples which are compared with a somewhat larger number exposed to  $^{60}\text{Co}$  gammas. Standard deviations of these ratios are typically 5%. In addition there is an uncertainty in the proton dose of about 5% due primarily to difficulty in measuring the diameter of the pin holes used to measure the proton fluence.

The measured response ratios are listed in Table II as a function of ion energy. The linear energy transfer (LET), which is the average stopping power over the ion range, is also shown as it is expected that the response ratio may be LET dependent. The following relations between response and LET can be derived from Table II for the phosphor  $\text{CaF}_2\text{:Mn}$ . The response to 2 MeV protons is about 30% less than that for gammas - a 30% reduction for about a factor of 100 increase in LET. There is a further decrease by about a factor of two as the average LET is increased by about 60% with decreasing proton energy. An additional reduction in response of about 20% is observed as the LET is increased by a factor of three by using 4.3 MeV alphas, and a further reduction of about 30% occurs when the alpha energy is reduced to 1.4 MeV, involving only a small increase in LET.

It should be noted that these are average LET values for the tabulated initial energies as the ions are reduced from their initial energies to zero energy in the phosphor. The actual LET is a function of the ion energy during the slowing-down process as seen in Fig. 2. Also, if the effect on response is one due to ion density, such as charge recombination or trap saturation (Ref. (6)) any LET dependence may be related to the square of the LET, or at least to the LET to some power greater than one. Bearing these facts in mind, it can be concluded that the LET effect for this phosphor is probably small for protons of energy greater than 1 MeV where the LET is less than  $200 \text{ MeV}/(\text{g}/\text{cm}^2)$  ( $60 \text{ keV}/\mu\text{m}$ ) as seen in Fig. 2 for  $\text{CaSO}_4$  which has approximately the same LET as  $\text{CaF}_2$ , and that any LET effect for protons with higher initial energies is due to the high LET values during the slowing-down process.

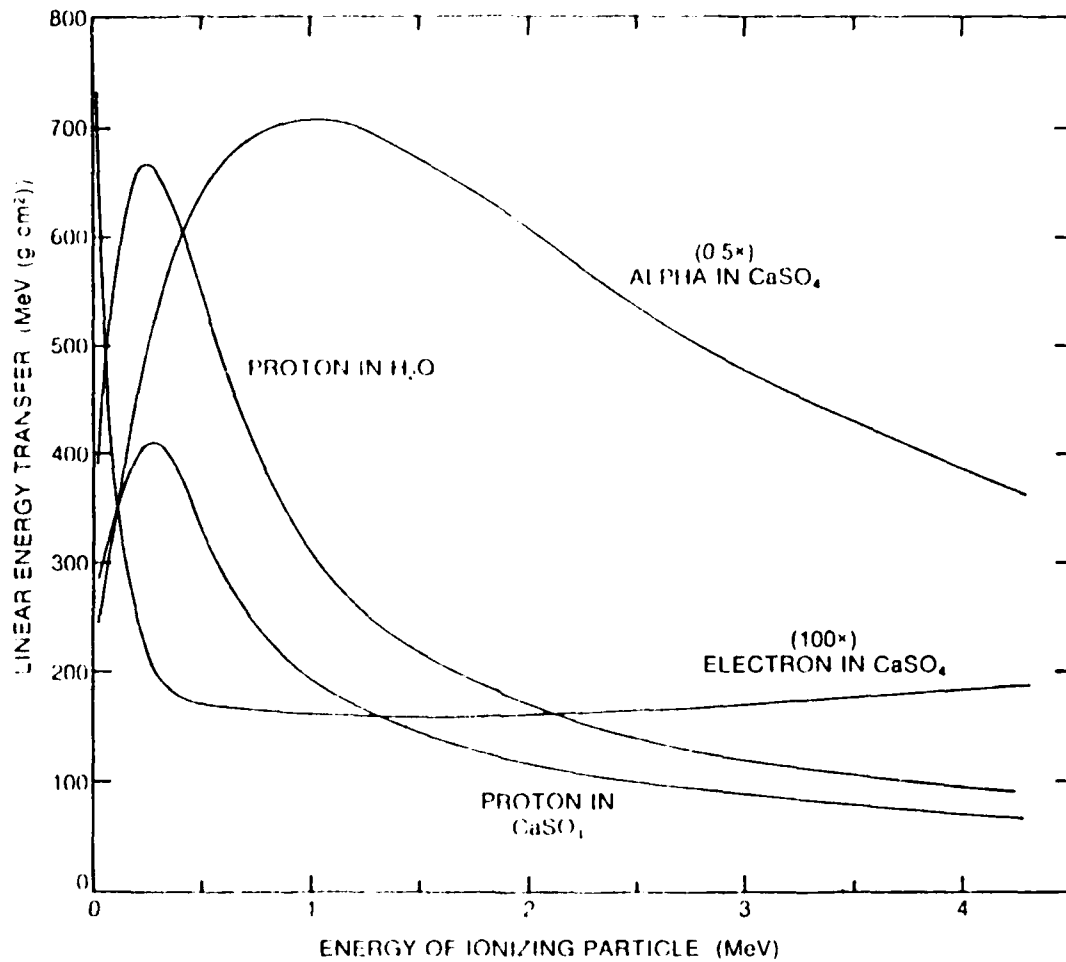


Figure 2: Linear energy transfer (LET) of electrons, protons and alphas in calcium sulphate and protons in water. For convenience of plotting, the LET has been divided by two for the alphas and multiplied by 100 for the electrons.

Table II

Response of  $\text{CaF}_2\text{:Mn}$  to Proton and Alpha Irradiations, Relative to  $^{60}\text{Co}$  Irradiation,  $\eta_p$  and  $\eta_\alpha$  for  $\text{CaF}_2\text{:Mn}$  Dosimeters Corrected for TL Absorption in the Samples. Types A and B are Polycrystalline Samples While Type S Refers to the Phosphor Powder in Silastic. The Approximate Ion Range and Average Linear Energy Transfer (LET) in  $\text{CaF}_2$  Are Also Listed.

| Ion    | Ion Energy (MeV) | Ion Range ( $\mu\text{m}$ ) | LET (keV/ $\mu\text{m}$ ) | Type: | Ion/Gamma Response Ratio |      |      |
|--------|------------------|-----------------------------|---------------------------|-------|--------------------------|------|------|
|        |                  |                             |                           |       | A                        | B    | S    |
| Proton | 2.00             | 35.0                        | 70                        |       | 0.69                     | 0.68 | 0.76 |
| "      | 1.00             | 12.0                        | 95                        |       | 0.52                     | 0.53 | 0.66 |
| "      | 0.90             | 10.0                        | 98                        |       | 0.53                     |      |      |
| "      | 0.80             | 8.5                         | 100                       |       | 0.52                     | 0.48 | 0.64 |
| "      | 0.64             | 6.5                         | 110                       |       | 0.46                     | 0.42 | 0.52 |
| "      | 0.37             | 3.5                         | 115                       |       | 0.38                     | 0.35 | 0.45 |
| Alpha  | 4.30             | 14.0                        | 320                       |       | 0.32                     | 0.30 | 0.39 |
| "      | 1.40             | 4.0                         | 360                       |       | 0.24                     | 0.20 | 0.22 |

Substantial changes in  $\eta_p$  and  $\eta_\alpha$  are seen in Table II for fairly small changes in LET. These occur with somewhat larger variations in ion range than in LET, and a relation between  $\eta$  and ion ranges is indicated. This could result from a reduction in sensitivity of the phosphor material near its surface to a depth less than the ranges of the higher-energy ions. Thus, it is difficult to separate an LET effect from a surface effect under many experimental conditions. The most valid conclusions on the LET effect of the ions on response can be drawn from the relative values of  $\eta$  for protons and alphas of equal ranges. The 4.3 MeV alphas have a range of 14  $\mu\text{m}$  and an average LET of 320 keV/ $\mu\text{m}$ ; protons of the same range have an energy of 1.1 MeV and an LET of 93 keV/ $\mu\text{m}$ . Comparison of the corresponding values of  $\eta$  in Table III shows that for this increase in LET the response is reduced by a factor of approximately 0.55 for the three  $\text{CaF}_2\text{:Mn}$  samples listed. A comparable relation is seen for the 1.4 MeV alphas and the low-energy protons of the same range.

### 3.3 Response Ratios $\eta_p$ of $\text{CaSO}_4:\text{Tm}$ and $\text{CaF}_2:\text{Dy}$ Relative to $\eta_p$ for $\text{CaF}_2:\text{Mn}$

Most of the proton irradiations of the  $\text{CaSO}_4:\text{Tm}$  samples were made with simultaneous irradiation of  $\text{CaF}_2:\text{Mn}$  samples, usually the type A polycrystalline dosimeters. Thus, the relative proton response of the two phosphors was made directly without having to rely on the measurement of the proton fluence. Since accompanying gamma irradiations were also done, the relative response for the two materials was measured and defined as:

$$\eta_{p21} = \eta_{p2}/\eta_{p1}, \quad (2)$$

where  $\eta_{p2}$  and  $\eta_{p1}$  are the proton/gamma sensitivity ratios, defined by Eq. (1).

Relative proton/gamma response ratios are listed in Table III for the two grades of  $\text{CaSO}_4:\text{Tm}$  powder investigated. These powders were embedded in Silastic to form dosimeters which were approximately 200  $\mu\text{m}$  in thickness. This exceeds the range of the 2 MeV protons which is about 70  $\mu\text{m}$  in Silastic. The proton/gamma sensitivity ratios for the  $\text{CaSO}_4:\text{Tm}$  are seen to decrease more with decreasing proton energy than the corresponding ratio for the  $\text{CaF}_2:\text{Mn}$  dosimeters. This decrease is not as great for the ultrafine powder. In fact, the responses of the two powders, which differ by a factor of two for gammas (see Table I), are about the same for the irradiations by the lower-energy protons. The large energy dependence of the response of the fine powder is probably related to the fact that the proton range (see Table II) is comparable to the diameter of the phosphor grains. A reduction in response with diameter of the phosphor grains and with proximity to the surface of the larger grains is indicated. The ultrafine powder is clearly the more suitable of these two powders for measurement of mixed neutron/gamma fields.

The response ratio for the  $\text{CaF}_2:\text{Dy}$  powder in Silastic follows closely to that of the ultrafine  $\text{CaSO}_4:\text{Tm}$  as the proton energy is decreased with slightly larger ratios being found for the  $\text{CaF}_2:\text{Dy}$ . Only the ultrafine powder was investigated for the latter phosphor.

TABLE III

Proton/Gamma Response Ratios for  $\text{CaSO}_4\text{:Tm}$  and  $\text{CaF}_2\text{:Dy}$  Relative to  $\text{CaF}_2\text{:Mn}$  Dosimeters (Type A). Relative Response Ratios for Both Fine and Ultrafine (UF)  $\text{CaSO}_4\text{:Tm}$  in Silastic Are Listed. The Actual Ratios Can Be Derived by Multiplying the Values in This Table by the Ratios for  $\text{CaF}_2\text{:Mn}$  (Type A) in Table II.

| Proton Energy (MeV) | <u>Relative Proton/Gamma Response Ratio</u>        |  |   |
|---------------------|--|--|---|
|                     | <u><math>\text{CaSO}_4\text{:Tm}</math> (Fine)</u> | <u><math>\text{CaSO}_4\text{:Tm}</math> (UF)</u> | <u><math>\text{CaF}_2\text{:Dy}</math> (UF)</u> |
| 2.0                 | 1.10   | 1.17   | 1.17  |
| 1.0                 | 0.98   | 1.27   | 1.33  |
| 0.8                 | 0.92   | 1.08   | 1.18  |
| 0.64                | 0.68   | 1.04   | 1.08  |
| 0.37                | 0.38   | 0.89   | 0.94  |

### 3.4 Ion/Gamma Response Ratios for Phosphor Powders in Silastic

The proton/gamma response ratios measured for the phosphor powders in Silastic are derived from Tables II and III by multiplying the relative ratios by the ratio for the Type A  $\text{CaF}_2\text{:Mn}$  dosimeters. The resulting values are listed in Table IV along with the alpha/gamma response ratios for these phosphors.

The ion/gamma response ratios for these powders follow much the same pattern as for the polycrystalline dosimeters discussed earlier. The decrease in  $\eta_p$  with decreasing proton energy indicates a decrease in  $\eta_p$  with increasing LET, but there is little difference between  $\eta_p$  for the lowest-energy protons and  $\eta_\alpha$  for the 4.3 MeV alphas, despite the fact that the LET for these alphas is approximately three times as large as for the protons. Also, there is a larger difference in  $\eta_\alpha$  for the two energies for which the average LET's are almost the same. This difference is observed for all four powders and is greatest for the coarser  $\text{CaSO}_4\text{:Tm}$  powder.

As for the polycrystalline  $\text{CaF}_2\text{:Mn}$ , comparison of some of the values of  $\eta$  in Table IV, indicates that some of the response variations are more related to ion range than to LET. As suggested earlier, this could result from non-uniform sensitivity within the phosphor. For phosphor particles of diameter comparable to the range of the ions, relatively low sensitivity near the surfaces would lead to reduced response for short-range ions. This could explain the drop in response with energy of the less fine  $\text{CaSO}_4\text{:Tm}$ , but it is hard to accept that such a large variation in response could result from a surface effect of the phosphor in the case of the ultrafine powders where the dimensions of the phosphor grains are generally considerably less than the range of the ions. The manner in which the ultrafine powders are embedded in the Silastic could lead to erroneous measured values of  $\eta$ . Possibly, surface tension of the Silastic in its liquid form could prevent the phosphor particles from being uniformly distributed at the outer surface of the mixture. A surface layer of Silastic would have the effect of reducing response particularly for the short-range ions.

For alphas and gammas of equal ranges, a somewhat larger LET effect on the response is seen for the  $\text{CaSO}_4\text{:Tm}$  than for the  $\text{CaF}_2$  phosphors. The response of the ultrafine  $\text{CaSO}_4\text{:Tm}$  in Silastic was found to be approximately 40% as large when measured with the 4.3-MeV alphas as for 1.1-MeV protons. Both these particles have ranges of  $14\text{ }\mu\text{m}$  but the average LET for these alphas is approximately 3.4 times as large as for the protons.



TABLE IV

Ion/Gamma Response Ratios for Phosphor Powders in Silastic

| <u>Ion</u> | <u>Energy<br/>(MeV)</u> | <u>CaF<sub>2</sub>:Mn (UF)</u> | <u>CaF<sub>2</sub>:Dy (UF)</u> | <u>CaSO<sub>4</sub>:Tm (UF)</u> | <u>CaSO<sub>4</sub>:Tm (Fine)</u> |
|------------|-------------------------|--------------------------------|--------------------------------|---------------------------------|-----------------------------------|
| Proton     | 2.00                    | 0.76                           | 0.81                           | 0.79                            | 0.80                              |
| "          | 1.00                    | 0.66                           | 0.69                           | 0.65                            | 0.54                              |
| "          | 0.80                    | 0.64                           | 0.61                           | 0.56                            | 0.48                              |
| "          | 0.64                    | 0.52                           | 0.50                           | 0.52                            | 0.33                              |
| "          | 0.37                    | 0.45                           | 0.36                           | 0.34                            | 0.12                              |
| Alpha      | 4.30                    | 0.39                           | 0.38                           | 0.27                            | 0.26                              |
| "          | 1.40                    | 0.22                           | 0.16                           | 0.09                            | 0.05                              |

3.5 Alpha/Gamma Response Ratios for Loose Phosphor Powders

While it was not practical to irradiate loose phosphor powders in the proton beam some measurements were made of the response of these powders to alphas and gammas. The gamma irradiations were done with the powders in glass vials using a <sup>60</sup>Co source. Factors of 0.85 and 0.82 rad/R, which are equivalent to 8.5 and 8.2 nJ/(mg R), were used for the CaSO<sub>4</sub>:Tm and CaF<sub>2</sub>:Mn powders, respectively. Small samples of the powders were spread over the heating planchet to avoid self absorption during reading.

A different procedure was required for the alphas. A relatively thick layer of powder was packed onto the heating planchet to ensure total absorption of the alphas in the sample. A defined area of this sample was exposed to the alphas under vacuum. Following exposure the planchet was transferred to the reader with care being taken not to disturb the sample. For some of the ultra-fine powders it was found necessary to dampen the sample

with a liquid (ethanol was used for the  $\text{CaSO}_4$  and water for the  $\text{CaF}_2$ ) to obtain a compact layer which would stay in place. These were dried before irradiation and no effect on the TL properties was observed as a result of handling the samples in this manner.

Because of the range of the alphas, the alpha irradiations affect the phosphor samples to a depth of approximately  $4 \text{ mg/cm}^2$ . This is comparable to the sample thicknesses read for the gamma irradiated powders. TL absorption in this thickness should be small. However, a substantial fraction of the TL measured by the reader is due to reflection from the planchet or from the phosphor substrate in the case of the alpha irradiations. Some gamma-irradiated samples were found to give equal responses when read on the planchet directly or on a layer of unirradiated  $\text{CaF}_2\text{:Mn}$ . From this it can be concluded that the two surfaces have nearly equal reflectivities so that the efficiency for reading the gamma- and alpha-irradiated samples should be the same.

Measured alpha/gamma response ratios  $r_\alpha$  are listed in Table V for the ultrafine powders of the three TL materials investigated and also for some coarser powders of the same phosphors.

TABLE V

Alpha/Gamma Response Ratios for Loose Phosphor Powders

| Alpha<br>Energy<br>(MeV) | CaF <sub>2</sub> :Mn |      |      | CaF <sub>2</sub> :Dy |      | CaSO <sub>4</sub> :Tm |     |
|--------------------------|----------------------|------|------|----------------------|------|-----------------------|-----|
|                          | Coarse               | Fine | UF   | Coarse               | UF   | Fine                  | UF  |
| 4.3                      | 0.55                 | 0.8  | 0.9  | 0.35                 | 0.70 | 0.5                   | 1.0 |
| 1.4                      | 0.4                  | 0.7  | 1.05 | 0.15                 | 0.6  | 0.3                   | 0.8 |

The coarse  $\text{CaF}_2\text{:Mn}$  and  $\text{CaF}_2\text{:Dy}$  powders contained large crystals up to about  $100 \mu\text{m}$  in diameter, while the ultrafine powders for these phosphors contained only particles less than about  $1 \mu\text{m}$ . These UF powders are probably slightly finer than those used in the Silastic dosimeters, while the fine and UF  $\text{CaSO}_4\text{:Tm}$  powders are from the same separations as were used in the Silastic dosimeters for that phosphor. The fine  $\text{CaF}_2\text{:Mn}$  powder contained grains up to a few  $\mu\text{m}$  in diameter.

Reduction in the size of these phosphor particles leads to a reduction in sensitivity to gammas. The fine and UF  $\text{CaF}_2\text{:Mn}$  have approximately 1/3 and 1/6 the sensitivity to gammas found for the coarse powder, the UF  $\text{CaF}_2\text{:Dy}$  has approximately 1/10 the gamma sensitivity of the coarse powder, and the UF  $\text{CaSO}_4\text{:Tm}$  has about 1/10 the gamma sensitivity of the fine powder. Combining these reductions with the ratios in Table V shows that the response to alphas is also reduced by particle-size reduction but to a lesser extent than the response to gammas.

Reduction in response with particle size and with alpha range (or energy) support the proposal of reduced response at the phosphor-particle surface. But the near-unity values of  $\eta_\alpha$  for the ultrafine  $\text{CaF}_2\text{:Mn}$  and  $\text{CaSO}_4\text{:Tm}$  indicate that the effect of LET on the response of these phosphors in the form of ultrafine powders (in no solid matrix) is small. This is in contradiction of the observations for these powders in Silastic as summarized in Table IV.

#### 4. Summary and Conclusions

The proton/gamma and alpha/gamma response ratios have been measured for polycrystalline  $\text{CaF}_2\text{:Mn}$  dosimeters and for  $\text{CaF}_2\text{:Mn}$ ,  $\text{CaF}_2\text{:Dy}$  and  $\text{CaSO}_4\text{:Tm}$  TL powders. The reduction in sensitivity to 1 and 2 MeV protons and 4.3 MeV alphas relative to  $^{60}\text{Co}$  gammas has been attributed to reduction in light-conversion efficiency with increasing LET of the ionizing particles. However, further reduction in the measured value of  $\eta$  as the ion energy is decreased appear to be more related to the ion range than to the LET. A reduction in sensitivity near the surface of the phosphor has been proposed to account for this effect. Reductions in  $\eta$ , attributed in the literature to LET, may be due, in part, to these surface effects.

For a neutron/gamma TLD made sensitive to neutrons by embedding phosphor powder in polyethylene, the value of  $\eta_p$  (about 0.8 at 2 MeV) is large enough to provide good neutron response for high-energy neutrons, as found in Ref (4). The reduction in response, observed as the proton energy is reduced below 1 MeV, can account for the reduced neutron response for low-energy neutrons, as found in Ref (4), whether this is due to a true reduction in  $\eta_p$  because of LET effects or to a reduced response at the phosphor surface.

Of the three phosphor powders investigated the  $\text{CaF}_2\text{:Mn}$  was found to be marginally better than  $\text{CaF}_2\text{:Dy}$  and  $\text{CaSO}_4\text{:Tm}$  at low proton energies. Two size gradations of  $\text{CaSO}_4\text{:Tm}$  powder were investigated, and a comparison of  $\eta_p$  for these show an improvement in relative response to low-energy protons by using the ultrafine grade. This emphasizes the importance of using a very fine powder in the gamma/neutron TL dosimeter.

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Proton/gamma and alpha/gamma response ratios have been measured for very fine powders of the thermoluminescent phosphors  $\text{CaF}_2\text{:Mn}$ ,  $\text{CaF}_2\text{:Dy}$  and  $\text{CaSO}_4\text{:Tm}$ . This has been done as part of an evaluation of these phosphors for use in a combined neutron/gamma thermoluminescent dosimeter which is made neutron sensitive by embedding the phosphor powder in an hydrogenous medium. The  $\text{CaF}_2\text{:Mn}$  phosphor has been found to be marginally better than the other two for this purpose. Reduction in the measured proton/gamma response ratio with decreasing proton energy has been attributed primarily to relatively low response near the surfaces of the phosphor particles.

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